High Resolution Simulations of Particle Sulfate Formation in Lake Breeze Fronts: Process Tracking and Implications for Forecasting.

P.A. Makar[1], C. Stroud[1], J. Zhang[1], D. Sills[2], W. Gong[1], M.D. Moran[1], D. Sills[2], S. Gong[1], K. Hayden[1], J. Brook[1], C. Mihele[1], J. Liggio[1], S.M. Li[1], J. Murphy[3], J. Abbatt[3], J. Slowik[3], G. Evans[3]

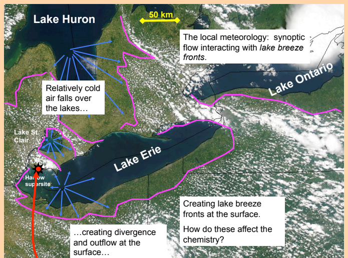
1. Air Quality Research Division, Environment Canada, 2. Cloud Physics and Severe Weather Research, Environment Canada, 3. University of Toronto

BAQS-Met 2007

Measurement and modelling study with the aim of examining the interactions between long-range transport, local circulation (lake breezes) and local emissions/chemistry, in S.W. Ontario.

→What is the impact of the local circulation and emissions on local air-quality (versus long-range transport)?

→How do trace gases and particles evolve downwind of a large, midlatitude urban and industrial centre (Detroit)?



AURAMS

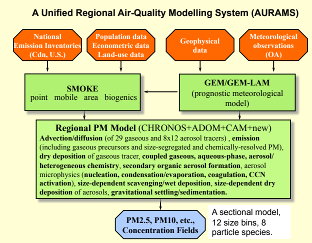
A Unified Regional Air-quality Modelling System was used to simulate air pollution during the 27 day period June 17 to July 11, 2007.

→2 levels of nesting in the meteorological driver.

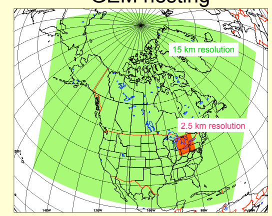
→3 levels of nesting in the AQ model.

→Mass balance trackers switched on for ozone and PM2.5 bins:

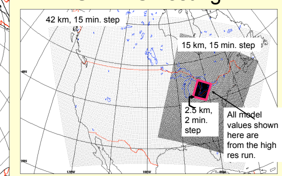
Tracking change in mass across every process in the model (Dmass/Dt)



GEM nesting



AURAMS nesting



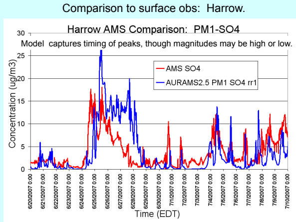
28 vertical levels (14 below 2km agl).

Particle Sulphate

- The AURAMS code includes switches for *mass-trackers*: the mass change (after-before) for each operator, every particle species and bin size, can be used to determine the relative importance of:
  1. *Gas-phase production and loss.*
  2. *Advection*
  3. *Diffusion, surface emission, and deposition.*
  4. *Net transport (2. + 3.).*
  5. *Total particle microphysics and chemistry.*
     - *Condensation.*
     - *Coagulation.*
     - *Nucleation.*
     - *In-Cloud Processes + Inorganic Heterogeneous Chemistry.*
     - *Below-Cloud processing.*
     - *Settling and dry deposition.*
- Surface contours, cross-sections, and 3D mapping of these mass tracking fields help show how the particulate matter forms, in the model, and gives hypotheses for testing against the observations.
- Here, the focus will be on PM1 SO4 simulations and comparison to observations.

PM_1-SO_4 at Harrow

Comparison to surface obs: Harrow.



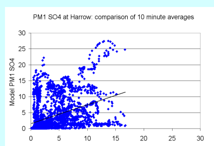
PM_1-SO_4 comparison with AMS (10 minute averages) at Harrow, entire intensive period

R: 0.3964

Best fit: model = 0.5933 obs + 1.556

Mean bias: -3.243E-03 ug/m^3

Mean error: 3.376 ug/m^3



Mass tracking of Particle Sulfate at Harrow

(June 24th 6pm to June 29th, midnight).

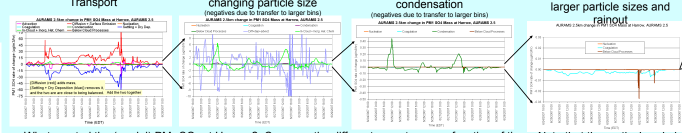
Mostly Transport

A little bit of in-cloud and heterogeneous chemistry changing particle size (negatives due to transfer to larger bins)

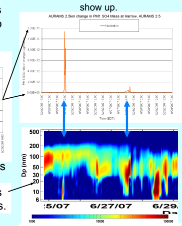
Small amount of H_2SO_4 condensation (negatives due to transfer to larger bins)

Very small amount of mass is lost due to coagulation to larger particle sizes and rainout

Then nucleation events start to show up.



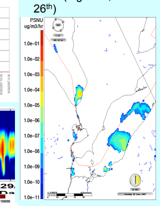
What created the (model) PM_1 SO_4 at Harrow? Compare the different operators as a function of time. Note that the vertical scale is getting smaller for each successive graph going from left to right!



Particle nucleation events compared to observations.

Harrow SPMS Observations courtesy Greg Evans, Cheol-Heon Geong, U of T.

Nucleation events predicted over the lakes (e.g. 18Z, June 26th)



Airborne AMS Measurements

NRC Twin Otter

Aircraft (over all flights):

R: 0.5541

Model = 1.029 Obs + 3.663

Mean Bias: 3.759

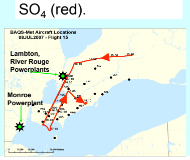
Mean Error: 4.592

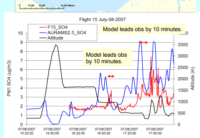
The Twin Otter measurements (and certain time periods within the surface measurements) suggest that the model PM1 SO4 is biased high.

→What can mass tracking tell us about the possible causes for the positive bias?

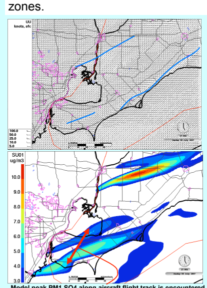
An example: Flight 15.

Top: flight track 15. Bottom: Altitude (black), model PM_1 SO_4 (blue), observed PM_1 SO_4 (red).



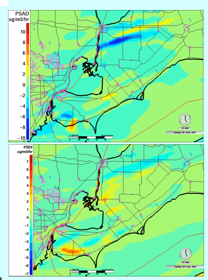


Top: Model wind fields, frontal convergence zones in blue. Bottom: PM_1 SO_4 concentrations at 815 m agl: power-plant plumes caught in lake-breeze frontal convergence zones.

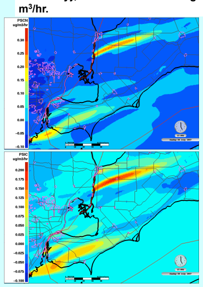


Model peak PM1 SO4 along aircraft flight track is encountered here.

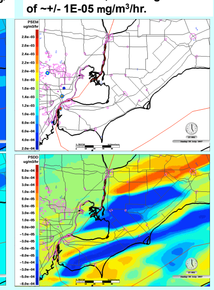
Net advection (top) and diffusion (bottom): mass transport on the order of 1 to 10 ug/m^3/hr.



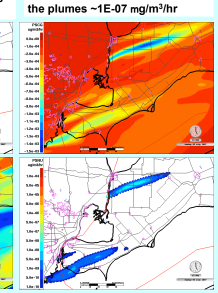
Top: Net PM_1 SO_4 mass change due to condensation; increases up to 0.3 mg/m^3/hr. Bottom: Net change due to {cloud processing + inorganic heterogeneous chemistry}, max increase 0.275 mg/m^3/hr.



Top: direct emissions of PM_1 SO_4; ~3E-03 mg/m^3/hr. Bottom: Particle settling: note that settling is decreasing one plume's mass within this altitude, but increasing that of another, with magnitudes of ~+/- 1E-05 mg/m^3/hr.



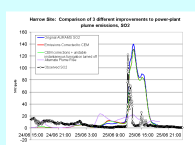
Top: Coagulation is shifting particle mass to larger size bins, ~1E-03 mg/m^3/hr. Bottom: particle nucleation increases mass in the smallest size bin in the plumes ~1E-07 mg/m^3/hr

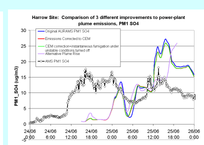


The mass tracking depicted at left suggests that the transport terms (advection, diffusion) have the largest impact on PM_1 SO_4 mass. Small errors in the transport direction (e.g. plume buoyancy height, wind speed and direction, strength of vertical diffusion) may therefore have a large impact on the PM_1 SO_4 concentration.

Investigation of different plume rise parameterizations at Harrow.

One possible reason for the positive bias in sulphate might relate to the plume rise (hence vertical placement of sulphate precursor SO_2). Try a few different methods of plume rise…





The model appears to capture the SO_2 arriving at Harrow from the Monroe power plant fairly well, but the model sulphate arrives late and peaks too high. Changing the initial mass distribution in the plume rise has a big effect on the SO_2, and delays the arrival of the main peak (but does not improve either SO_2 or PM_1 SO_4 simulations).

Conclusions (for a work in progress):

(1) Lake breeze convergence zones have a significant impact on particle sulphate concentrations (along with fortuitous power-plant locations!)

(2) Lake breeze circulation transports SO2 and PM_1 SO_4; local concentrations are often highest in lake-breeze convergence lines.

(3) High spatial and time resolution modelling is difficult: It's hard to get the $R^2 > 0.6$, slope = 1.0 behaviour of the coarser resolution version of the same model (which is usually compared to 24 averaged, one day in 3 or 6 network data).

(4) Small errors in plume placement have a large effect at high res!

(5) Mass tracking: local circulation has a big impact on predicted concentrations!

(a) Many processes (rainout, aqueous chemistry, condensation, etc.) strongest in convergence lines.

(b) Nucleation events strongest over lakes.

(c) The fastest rates of change are in lake-breeze fronts